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# Degradation and recovery of adhesion properties of deformed metal–polymer interfaces studied by laser induced delamination

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## Abstract

Adhesion properties of polymer coatings on metals are of great interest in various industrial applications, including packaging of food and drinks. Particular interest is focused on polymer–metal interfaces that are subjected to significant deformations during manufacturing process. In this work steel samples laminated with polyethylene terephthalate (PET) were subjected to uniaxial tensile deformations followed by annealing treatments. The measurements have demonstrated degradation of adhesion of the metal–polymer interface as the strain introduced by the deformation increased. Moreover, it was observed that within the geometry used in the experiments tensile deformations of the metal substrate introduced in-plane compressive stresses in the bulk of the coating. After applying a thermal treatment restoration of the adhesion has been achieved.

Laser induced delamination technique was used to monitor the adhesion properties. In this technique a coating is subjected to a series of infrared laser pulses with a stepwise increase of intensity. Upon increasing the laser pulse intensity, the pressure which is formed inside the blisters reaches a critical value, resulting in further delamination of the coating. To process the experimental data an elastic model was developed. From the analysis of the experimental data the critical stresses required for the delamination and the practical work of adhesion are derived. The model accounts for the compressive in-plane stress present in the coating of the deformed samples.

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**Keywords:** Coating; Adhesion; Polyethylene terephthalate; Residual stress; Elastic theory

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## 1. Introduction

In many industrial applications metal substrates coated with a polymer film are subjected to significant mechanical deformations, which cause degradation of adhesion properties of the polymer–metal interface. Various physical processes triggered by deformations can potentially influence the adhesion properties. For example, a tensile deformation of a metal substrate will introduce in-plane stresses in the polymer coating and may alter the actual contact area. Further, deformation increases the roughness of the metal surface in contact with the polymer coating and may lead to delamination of the adherent layer. The influence of the interface roughness due to plastic deformation of the metal substrate on the work of adhesion and on the interface energy with a glassy polymer is numerically studied in Ref. [1]. By performing a finite element analysis (FEA) it was

demonstrated that the interface energy decreases until the strain at yield of the polymer coating. After yielding as the polymer starts to soften macroscopically, the decreasing average stress levels result in a partial recovery of the interface energy at the interface. At higher strains, when macroscopic hardening develops the recovery of the interface stops and the interface energy decreases. In another study, FEA code and a simple beam theory were applied to analyze the changes in the interfacial strain energy release rate in the double cantilever beam (DCB) specimens caused by in-plane residual stresses [2]. The origin of the in-plane residual stresses in this case was the mismatch in thermal expansion coefficients between the polymer and the metal substrate. Both analytical solutions and FEM numerical simulations showed that the residual-stress-induced energy release rate is always higher for compressive residual stress than for tensile residual stress.

Currently various measuring techniques aimed at testing adhesion of polymer–metal interfaces are available [3,4]. However, not many experimental studies are reported in literature in which the effect of deformation on the adhesion of the interface

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is considered. In the experimental study reported in Ref. [5] effects of tensile deformation on adhesion were directly measured from the difference in the load versus elongation curves between metal–film/substrate and pure substrate structures. This method has been applied to study Cu(Cr)/polyimide structures.

Deformation of a metal substrate also introduces in-plane stresses in the polymer film. Such stresses can weaken the polymer–metal interface and in the extreme case can lead to spontaneous delamination. Effect of in-plane stresses on adhesion has been extensively covered in literature, although in most cases the origin of the in-plane stress is of a different nature [6,7]. Similar to deformation of a substrate, in-plane stresses can be introduced in a coating due to a mismatch in thermal expansion coefficients between the polymer and the metal substrate. Thus in-plane residual stresses of thermal origin were measured with the curvature based techniques [8–11], double cantilever test [2,12–14] and other less common experimental methods [15–17]. High compressive stresses are also found in coatings deposited on various substrates. In many cases these compressive stresses cause delamination and buckling of the film. In Ref. [18] the buckling instability condition was utilized for quantitative characterization of the adhesion of a carbon ion beam deposited layer on a quartz substrate.

In this work laser induced delamination technique was used to investigate the effect of a tensile deformation on the adhesion properties of the PET–steel interface. In this method nanosecond infrared laser pulses are used to form initial blisters at the polymer–metal interface [19]. Upon increasing the laser pulse intensity, the pressure which is formed inside the blisters reaches a critical value, resulting in further delamination of the coating. From the shape of the blister the adhesion properties of the interface are evaluated. Laser induced delamination test does not require any special sample preparation, which makes this method very suitable for monitoring the changes in the adhesion properties of the interface as the deformation of the specimen progresses.

To process the experimental data a simple elastic model was developed and presented in earlier publications [19,20]. Using this model the critical stresses required for delamination and the practical work of adhesion were derived. To account for possible plastic deformation and to validate the analytic model computer simulations using finite elements analysis with a mixed mode cohesive zone were carried out [20]. A fair agreement between the stress fields calculated with FEA code and those predicted by the elastic model has been demonstrated.

This elastic model, however does not account for the in-plane stresses which can be present in the polymer film. In order to analyze the experimental data obtained on the deformed samples, when the in-plane stresses could not be neglected, the model was revised. The revised version of the model is presented in Section 2 of this work.

The experimental results reported in this work include a study on the adhesion properties of polyethylene terephthalate (PET)—interstitial free (IF) steel interface subjected to tensile deformation followed by annealing. The evolution of the practical work of adhesion was monitored by means of the laser induced delamination technique.

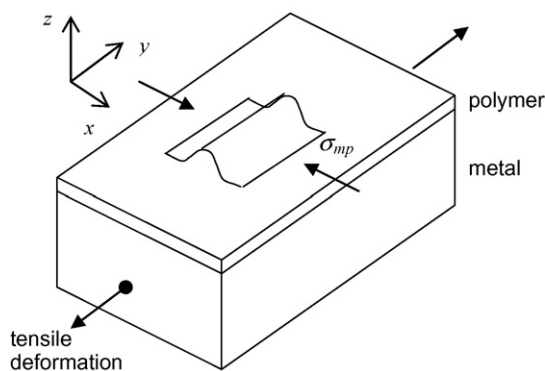


Fig. 1. Schematic representation of the geometry of the experiment. The orientation of the blisters with respect to the direction of the tensile deformation of the metal substrate is shown.

## 2. Theoretical model

Standard thin plate model based on the Kirchhoff assumptions [21] does not consider any stretching or compression in the mid-plane of the coating. Only bending of the mid-plane contributes to the strain energy. Such assumption is not correct if in-plane stresses are present in the film that arise by deformation or thermal treatment. The model presented below describes the formation of a blister at the polymer–metal interface in the presence of a compressive mid-plane stress. As will be shown in Section 3, only this kind of mid-plane stresses are expected in the samples used in the measurements. To facilitate the measuring procedure the blisters were formed in a cylindrical shape. As demonstrated in Fig. 1, the orientation of the blisters was chosen in such a way, that the cylindrical axis of the blister is perpendicular to the direction of the mid-plane stress.

A cylindrical blister aligned along the  $y$ -axis can be described as a thin plate clamped along the boundaries parallel to the  $y$ -axis (see Fig. 1). The governing equation for deflection  $w$  of a plate under normal uniform pressure  $p$  in 2D in presence of the mid-plane stress  $\sigma_{mp}$  is written as [21,22]:

$$\frac{d^4 w}{dx^4} = \frac{p}{D} - \frac{\sigma_{mp} t}{D} \frac{d^2 w}{dx^2}, \quad (1)$$

where  $D = Et^3/12(1 - \nu^2)$  is the flexural rigidity,  $E$  is the modulus of elasticity,  $\nu$  the Poisson's ratio and  $t$  is the film thickness. The blisters are overpressurized and the pressure excess over the atmospheric pressure  $p_{atm}$  is denoted by  $p$ . Then the absolute pressure inside the blister is  $p_{abs} = p_{atm} + p$ . The second term in the right-hand part of Eq. (1) accounts for the compressive stress. This contribution is weighted with the second derivative of the deflection which is the bending curvature of the film. This curvature determines the projection of the mid-plane stresses onto the vertical direction ( $z$ -axis), which contribute to the bending of the film. In case of tensile mid-plane stresses this term appears with a positive sign. Clamped, or built-in, boundary conditions are used:

$$w = 0, \quad w' = 0, \quad \text{at } x = -\frac{a}{2} \text{ and } x = \frac{a}{2}, \quad (2)$$

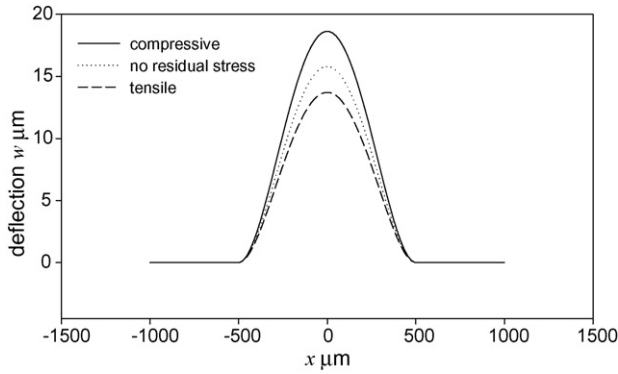


Fig. 2. Blister profiles calculated for the cases of a compressive mid-plane stress ( $\sigma_r = 1$  MPa), zero mid-plane stress, and tensile mid-plane stress ( $\sigma_r = 1$  MPa). PET film thickness is  $30 \mu\text{m}$  and the blister overpressure is  $p = 0.3 \times 10^5$  Pa.

where  $a$  is the dimension of the blister along the  $x$ -axis. The solution for this boundary value problem is

$$w(x) = \frac{pa}{2D\gamma^3} \frac{\cos(\gamma x) - \cos(\gamma(a/2))}{\sin(\gamma(a/2))} + \frac{p}{2D\gamma^2} \left( x^2 - \left( \frac{a}{2} \right)^2 \right), \quad (3)$$

where

$$\gamma = \sqrt{\frac{\sigma_{\text{mp}} t}{D}}. \quad (4)$$

Consequently, the blister height is:

$$H = p \left( \frac{a}{2D\gamma^3} \frac{1 - \cos(\gamma(a/2))}{\sin(\gamma(a/2))} - \frac{1}{2D\gamma^2} \left( \frac{a}{2} \right)^2 \right) \quad (5)$$

In the limiting case of  $\gamma \rightarrow 0$ , Eq. (3) converges to the earlier obtained solution for the film deflection without the mid-plane stresses [19]:

$$w(x) = \frac{pa^4}{24D} \left( \left( \frac{x}{a} \right)^2 - \frac{1}{4} \right)^2 \quad (6)$$

In Fig. 2 three blister profiles are compared: (1) in the presence of 1 MPa mid-plane compressive stress Eq. (3); (2) zero mid-plane stress Eq. (6); (3) in the presence of 1 MPa mid-plane tensile stress. It is demonstrated that compressive stresses make the blister higher, while tensile stresses make the blister lower.

In practice blisters have a finite length. It is convenient to introduce the blister length  $b$ , measured in the  $y$ -direction. The volume of the blister is obtained by integrating Eq. (3):

$$V = pf(a)b, \quad (7)$$

where

$$f(a) = \frac{a}{D\gamma^4} - \frac{a^2}{2D\gamma^3} \frac{\cos(\gamma(a/2))}{\sin(\gamma(a/2))} - \frac{2}{3} \frac{1}{D\gamma^2} \left( \frac{a}{2} \right)^3. \quad (8)$$

Note that in reality the blister is also clamped at the boundaries  $y=0$  and  $y=b$ . However, if  $b \gg a$  the error introduced is negligible.

The elastic strain energy has two contributions: bending energy  $U_b$  and strain energy stored in compression of the mid-

plane  $U_{\text{mp}}$ . The bending energy can be calculated as follows [22]:

$$U_b = \frac{Db}{2} \int_{-a/2}^{a/2} (w''(x))^2 dx = p^2 q_b(a)b \quad (9)$$

where

$$q_b(a) = \frac{a^2}{8D\gamma^3} \frac{\cos(\gamma(a/2))\sin(\gamma(a/2)) + \gamma(a/2)}{\sin^2(\gamma(a/2))} - \frac{a}{2D\gamma^4}. \quad (10)$$

The strain of the mid-plane can be written as follows [22]:

$\varepsilon = \varepsilon_\sigma + (1/2)(w')^2$ . The first term represents the original compression of the film due to the stress  $\sigma_{\text{mp}}$ . The deflections of the film considered in this work are small and far from the buckling condition. Therefore, neither bending nor delamination relieves this stress, and we assume that the strain energy associated with this term does not change during delamination. The second term, however affects the blister height and the strain energy associated with this term should be included in the energy balance condition required for delamination:

$$U_{\text{mp}} = \frac{\sigma_{\text{mp}} t b}{2} \int_{-a/2}^{a/2} (w')^2 dx = p^2 q_{\text{mp}}(a)b, \quad (11)$$

where

$$q_{\text{mp}}(a) = \frac{3}{8} \frac{a^2}{D\gamma^3} \frac{1}{\tan(\gamma(a/2))} + \frac{1}{D\gamma^2} \left( \frac{a}{2} \right)^3 \times \left( \frac{1}{3} + \frac{1}{2 \sin^2(\gamma(a/2))} \right) - \frac{a}{D\gamma^4} \quad (12)$$

The condition for delamination has the following form:

$$-(dF + dU + \delta A) \geq G dS. \quad (13)$$

The left-hand part presents the total (or global) energy release rate, which includes the following terms:  $dF = -(p_{\text{atm}} + p) dV$  is the decrease of the Helmholtz free energy of the gas inside the blister,  $dU = dU_b + dU_{\text{mp}}$  is the strain energy release rate, which includes two contributions: bending of the blister cap and the mid-plane compression, and  $\delta A = p_{\text{atm}} dV$  is the work produced against the outside pressure  $p_{\text{atm}}$ .  $G$  is the practical work of adhesion and  $dS = b \times da$  is the delaminated area. The measured practical work of adhesion  $G$  comprises the thermodynamic work of adhesion and the irreversible plastic dissipation part. It has been previously demonstrated that in the laser induced delamination test the stress fields introduced in the bulk of the polymer do not exceed the yield stress of the polymer [20]. Therefore, the contribution of the plastic deformations is only limited to the crack tip area. The film stops to delaminate when the total energy release rate is equal to the practical work of adhesion. Further in the paper no distinction is made between both of these concepts.

During delamination the blister volume increases and assuming that the amount of gas inside the blister stays constant

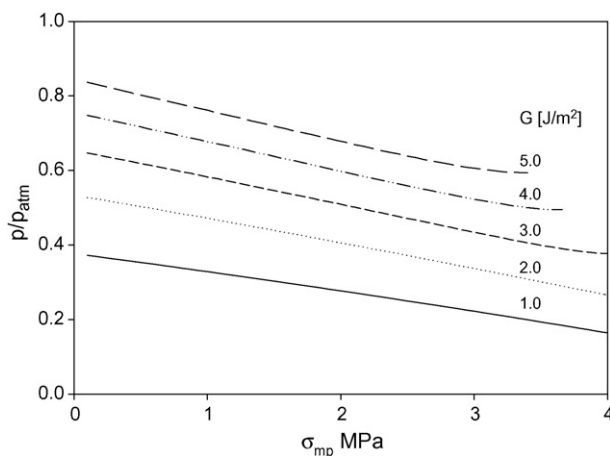


Fig. 3. Blister overpressure required to delaminate the PET film (30  $\mu\text{m}$ ) for the indicated values of the work of adhesion as a function of mid-plane compressive stress.

$(p_{\text{atm}} + p)V = \text{const.}$ , the blister overpressure  $p$  drops:

$$\frac{\partial p}{\partial a} = -\frac{p(p_{\text{atm}} + p)}{(p_{\text{atm}} + 2p)} \frac{f'}{f} \quad (14)$$

The differentials  $(dU/da)$  and  $(dV/da)$  are found:

$$\frac{dV}{da} = \left(\frac{\partial V}{\partial a}\right) + \left(\frac{\partial V}{\partial p}\right) \left(\frac{\partial p}{\partial a}\right) = \frac{p^2}{(p_{\text{atm}} + 2p)} f', \quad (15)$$

$$\frac{dU}{da} = \left(\frac{\partial U}{\partial a}\right) + \left(\frac{\partial U}{\partial p}\right) \left(\frac{\partial p}{\partial a}\right) = p^2 q' - 2 \frac{p(p_{\text{atm}} + p)}{(p_{\text{atm}} + 2p)} q \frac{f'}{f}. \quad (16)$$

The sum of two contributions to the strain energy are denoted as  $U = U_b + U_{\text{mp}}$  (and  $q = q_b + q_{\text{mp}}$ ). Combining Eqs. (13), (15) and (16) the final expression for the practical work of adhesion in terms of the functions  $f$ ,  $q$  and their derivatives in respect to the blister width  $a$  is obtained:

$$G = \frac{p^2 f'}{(p_{\text{atm}} + 2p)} \left( p + 2(p_{\text{atm}} + p) \frac{q}{f} \right) - p^2 q' \quad (17)$$

It should be noted that the total energy release rate has two driving forces for delamination: gas pressure inside the blister and the compressive mid-plane stress (or, to be correct, its projection onto the  $z$ -axis). This statement is demonstrated in Fig. 3. In this figure the overpressure required to delaminate the coating which is characterized with the indicated work of adhesion, decreases if the mid-plane compression stress increases.

### 3. Experimental method

In the laser induced delamination technique presented in Ref. [19] a coating under study is subjected to a series of laser pulses with stepwise increase of intensity. Every shot is carried out through a mask resulting in a formation of a cylindrically shaped blister. Such geometry is chosen to facilitate the measuring procedure of the blister profile. Upon increasing the laser pulse intensity, the pressure inside the blister reaches the critical value, resulting in further delamination. The measured blister profiles

are fitted to the model presented below in order to calculate the stresses in the film and the practical work of adhesion. In some experiments by means of a different mask two parallel cylindrically shaped blisters are formed. The strip of film between the blisters is not exposed to the laser irradiation and delaminates only when the pressure inside the blisters reaches the critical value.

In the experiments we used the infrared Nd:YAG laser NL303HT from EXPLA with the maximum pulse energy of 800 mJ and 5 ns pulse duration. The blister profiles were measured with the stylus Perthometer S2 from Mahr. Samples under study were obtained from CORUS and presented 30  $\mu\text{m}$  layer of PET on interstitial free steel substrate. There was a few nanometers chromium-oxide layer between the steel and the polymer. Blister profiles were measured with the stylus profiler Perthometer S2 from Mahr.

Tensile deformation of the samples was carried out with the material testing system MTS 810. Anneals of the samples were performed out in a hot air oven.

### 4. Experimental results and discussion

The objective of this experimental study is to investigate the influence of tensile deformation and subsequent annealing on the adhesion properties of the polymer–metal interface. First the reference samples were measured to obtain the original value of the practical work of adhesion. The results are presented in Fig. 4. The axes of the plot are the blister width  $a$  and the height  $H$ . Every contour line represents the critical blister dimensions at which delamination of an interface characterized by the indicated practical work of adhesion (in  $\text{J/m}^2$ ) takes place. The critical dimensions are calculated with Eq. (17), with zero mid-plane stress ( $\gamma \rightarrow 0$ ). As the intensity of the laser pulse increases a blister height  $H$  and width  $a$ , follow the following trend. At low intensities, when no delamination occurs, the width of the blister is constant and is equal to the image of the opening in the mask projected on a sample. The height of the blister gradually increases with the intensity of the laser beam. As the laser intensity increases the pressure inside the blisters rises and eventually the condition for delamination is satisfied. Both the blister width and the height increase following one of the contour lines given by Eq. (17). The blister dimensions obtained from the profile measurements on the reference sample are shown with the closed circles. Most of the data points covering a wide range of blister widths are bound between two contour lines corresponding to 2 and 3  $\text{J/m}^2$ .

The tensile samples for mechanical testing were cut with a laser. To control if the heating of the sample during the laser cutting affected the adhesion, one tensile sample after the laser cutting was measured before being deformed. The data points are shown with the open circles in Fig. 4 and demonstrate that the cutting procedure did not affect the adhesion properties of the interface.

The samples were subjected to tensile deformations with 5% and 10% strain. The schematic representation of the experiment geometry is given in Fig. 1. The deformation of the metal substrate is described by plane stress condition. Full relaxation of



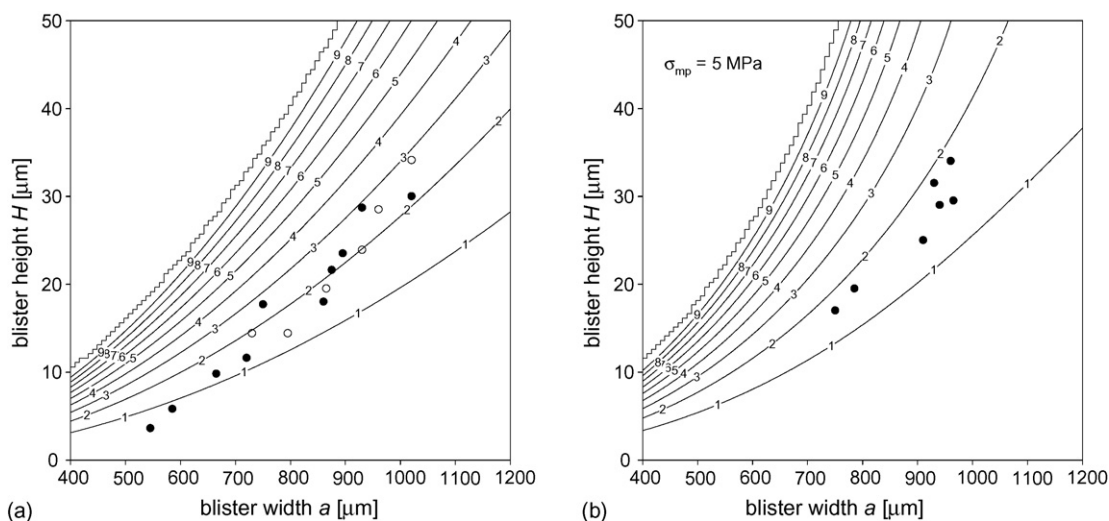


Fig. 4. Contour lines of constant work of adhesion are defined by Eq. (17). These lines present the blister critical dimensions, height and width, at which delamination of an interface characterized with the indicated work of adhesion takes place. Left figure corresponds to the case when no mid-plane stress is present ( $\gamma \rightarrow 0$ ). Right figure corresponds to the case when 5 MPa compressive stress acts perpendicular to the blister cylindrical axis. Experimental data is presented by symbols. Left: undeformed samples; right: the sample subjected to 5% strain tensile deformation.

strain in the metal substrate in the direction perpendicular to the direction of applied stress is accompanied by only partial strain relaxation in the polymer film, because of the two orders of magnitude difference in the Young's modulus. Therefore, deformation of the polymer film is closer to plain strain condition. The blisters were oriented with their cylindrical axis parallel to the applied stress. Thus the tensile deformation of the sample resulted in a compressive mid-plane stress, making the blisters higher as compared to those of the reference sample. For the sake of comparison future work will also include the formation of the blisters in the direction perpendicular to the direction of applied stress, which will result in a tensile mid-plane stress, making the blisters smaller. Both cases are demonstrated in Fig. 2.

In order to calculate the practical work of adhesion according to Eq (17), one needs to know the compressive mid-plane stress  $\sigma_{mp}$ . The following procedure was used to define this stress. First by using the reference sample it was estimated how much gas is generated inside the blister at different laser pulse intensities. The amount of gas is calculated as  $(p_{atm} + p)V$ , where Eqs. (5) and (7) were used to determine the blister pressure  $p$  and volume  $V$ , respectively. It was assumed that no mid-plane stress is present in the undeformed sample ( $\gamma \rightarrow 0$ ). The results are presented in Fig. 5 with open circles and demonstrate a linear dependency. If the same procedure is carried out for the deformed samples, the data points appear higher than those for the reference sample, as shown by closed circles for the 5% deformed sample. In all experiments, however, the laser pulse intensity was varied with the same step, and we assume that the same amount of gas was generated inside the blisters of the reference and deformed samples. The offset observed for the deformed sample is due to presence of the mid-plane compressive stress. By adjusting the value of the mid-plane compressive stress  $\sigma_{mp}$  the offset can be removed as shown in Fig. 5. The amount of gas generated per laser shot for the deformed sample is now more in accordance with the reference sample (shown by crosses). The fitted value of

the compressive mid-plane stress  $\sigma_{mp}$  for this sample is 5 MPa. In Fig. 4 the contour lines of constant practical work of adhesion are calculated with Eq. (17), for the case of the deformed sample (5% strain) with the obtained value of the compressive stress. From the comparison of this contour plot to the one in Fig. 4 for the reference, i.e. not deformed sample, one can observe that the lines are shifted to higher values of the blister heights. This shift is more pronounced as the compressive mid-plane stress increases. The same procedure was used to calculate the mid-plane stresses and the practical work of adhesion for all samples, subjected to tensile deformation and subsequent annealing.

The results are presented in Fig. 6. It is demonstrated that the practical work of adhesion decreases as the deformation of the samples progresses. The compressive mid-plane stress defined for each sample is indicated.

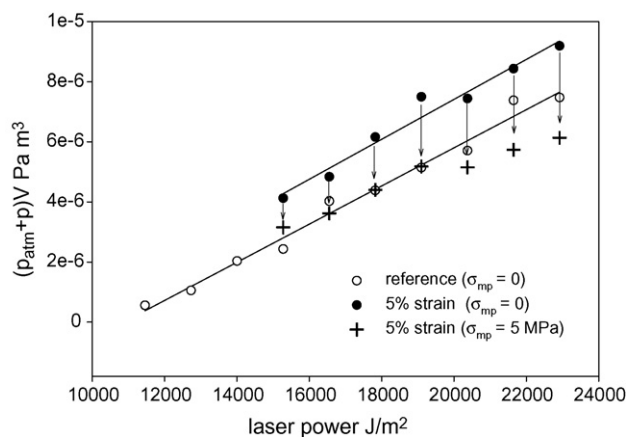


Fig. 5. Amount of gas generated inside the blisters as a function of the laser pulse intensity. Calculations carried out for the reference (not deformed) sample are shown with open circles and demonstrate a linear dependency. Closed circles and crosses represent the amounts of gas in the blisters for the deformed sample (5% strain) calculated with  $\sigma_{mp} = 0$  and  $\sigma_{mp} = 5$  MPa, respectively.

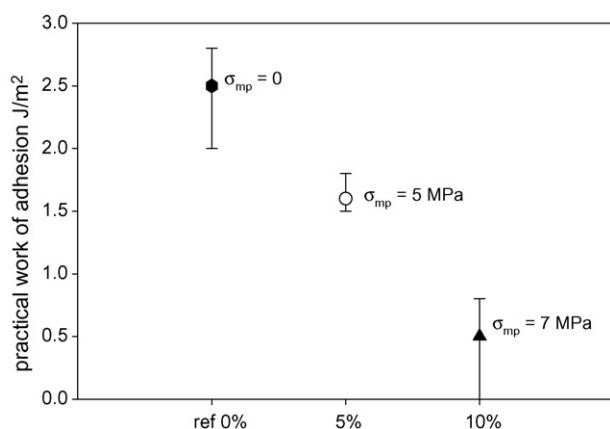


Fig. 6. Practical work of adhesion (or total energy release rate) measured for the reference sample and deformed samples (5% and 10% strain). The corresponding compressive in-plane stress used in the calculations is indicated.

As it has been mentioned, in the deformed samples there are two driving forces responsible for delamination: generated gas pressure inside the blisters and the mid-plane compressive stress. Gas pressure is introduced by the testing method while the compressive mid-plane stress already exists in the coating. This compressive stress makes the adhesion of the coating weaker. As shown in Fig. 3 one needs less gas in the blister to delaminate the coating in presence of a compressive stress, even if the value of the practical work of adhesion  $G$  does not change. In the extreme case, the compressive stress can lead to a spontaneous delamination and buckling. The measured decrease in the practical work of adhesion demonstrated in Fig. 6 is related to the weakening of the interface and has apparently a different nature: i.e. an increase of the roughness of the metal surface in contact with the polymer, degradation of the chromium oxide layer, which covered the steel surface prior to the deformation.

After deformation the tensile samples were annealed at 50 °C for 30 min and at 100 °C for 10 min in hot air. It was intended to anneal the samples at temperatures below and above the glass

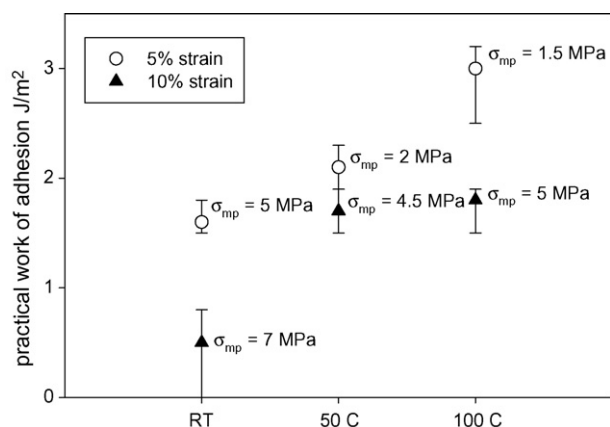


Fig. 7. Practical work of adhesion (or total energy release rate) measured for the deformed samples (5% and 10% strain) after anneals: 50 °C 30 min and 100 °C 10 min. The corresponding compressive in-plane stress used in the calculations is indicated.

transition temperature  $T_g$ , which in case of PET is 70–75 °C. The results are presented in Fig. 7. In all cases a recovery in the adhesion strength is observed. In the case of 5% deformation full recovery of the practical work of adhesion is attained. The compressive mid-plane stress has also decreased. In the case of 10% deformation the recovery of the adhesion is not complete. In the near future more research is planned to obtain a more in-depth understanding of the physical mechanisms responsible for the observed changes in the adhesion.

## 5. Conclusions

In this work the effect of tensile deformation followed by thermal annealing on the adhesion properties of polymer–metal interface was studied. The samples presented steel substrate laminated with polyethylene terephthalate. The measurements have demonstrated a decrease of the practical work of adhesion measured on the deformed samples. Moreover, within the geometry used in the experiments, tensile deformations of the metal substrate were shown to introduce in-plane compressive stresses in the bulk of the coating. After applying the thermal treatment a restoration of the adhesion properties has been observed. The adhesion of the interface was monitored with the laser induced delamination technique. To process the experimental data an elastic model was developed. The model accounted for the in-planes stresses present in the coating.

## Acknowledgement

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